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31223-78925 (B-9581)

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: Jean-Pierre Dath et al
Serial Number: 09/206,216
Filing Date: December 5, 1998
Examiner: T. Nguyen
Group Art Unit: 1764
Title: PRODUCTION OF OLEFINS

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APPEAL BRIEF

This is an appeal from the decision of the Primary Examiner finally rejecting claims 16-20. This Brief is submitted in triplicate with the statutory fee of \$320.00.

Real Party in Interest

The real party in interest is Fina Research, S.A., a Belgium corporation, as recorded by the assignment dated February 24, 1999, and recorded in the Patent and Trademark Office on March 22, 1999, at Reel/Frame 9843/0236.

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Related Appeals and Interferences

There are presently no appeals or interferences directly related to this appeal. However, commonly-assigned applications having the same inventive entity as, or a common inventor with this application and directed to catalytic cracking processes are the subjects of appeals as indicated below.

Application Serial No. 09/206,210; Notice of Appeal filed May 2, 2001

Application Serial No. 09/205,056; Notice of Appeal filed November 22, 2002

Application Serial No. 09/206,208; Notice of Appeal filed August 24, 2001

Application Serial No. 09/206,207; Notice of Appeal filed September 24, 2001

Application Serial No. 09/205,559; Notice of Appeal filed August 27, 2001

Application Serial No. 09/206,215; Notice of Appeal filed August 22, 2002

Application Serial No. 09/594,059; Notice of Appeal filed May 12, 2002

Status of Claims

The claims pending in this application upon filing of the Notice of Appeal were claims 1, 2, 7-10, 16-20, 24, 27-36. Upon entry of the amendment under 37 CFR 116 referred to below, claims 1, 2, 7-10, 12-14, 24 and 27-36 will be cancelled. Claim 16 will be cancelled and rewritten in independent form as claim 37 leaving claims 17-20 and 37 pending in this application. Claims 17-20 and claim 37 are appealed and reproduced in Exhibit A. No claim is allowed.

Status of Amendments

An Amendment under 37 CFR 1.116 is being filed concurrently with this Appeal Brief. It is appellants' understanding that this Amendment will be entered upon filing of this appeal brief.

Summary of the Invention

The claims on appeal are directed to a process for the cracking of olefins in an olefin-rich feedstock, having a maximum diene concentration of 0.1 wt. %, employing an MFI crystallate silicate catalyst having a silicon/aluminum atomic ratio of from 180-1,000 (page 14, second paragraph). The feedstock is supplied to the MFI crystallate silicate catalyst at an inlet temperature of 500-600°C. Other reaction conditions include an olefin partial pressure of 0.1-2 bars, and a space velocity (LHSV) of from 10-30 h⁻¹ (page 7, first paragraph). In one aspect of the invention, the cracking process, is specific to propylene and the effluent from the cracking process has a higher propylene content than the feedstock.

In a further aspect of the invention of the feedstock is treated to remove dienes from the feedstock prior to the cracking operation by selective hydrogenation of the dienes (page 7, second paragraph). The preliminary diene hydrogenation procedure is carried out at an inlet temperature of 40-200°C and a pressure of 5-50 bars, preferably 20-30 bars. The space velocity of the feedstock during the diene hydrogenation procedure is within the range of 2-5 h⁻¹ (paragraph bridging pages 19 and 20).

The MFI catalyst employed in appellants' invention is of a particular crystalline structure type as established by the Structure Commission of the International Zeolite Association as referred to on page 1 of appellants' specification. As stated there, ZSM-5 and silicalite are MFI-type crystalline silicates. Attached hereto as Exhibit B is a copy of pages 1-12 and 89-93 from the *Atlas of Zeolite Structure Types* by Meier et al, published by the Structure Commission of the International Zeolite Association in 1978. As indicated on page 93, ZSM-5 and silicalite (referred to in Footnote 1 by reference to Flanigen et al (1978)) are MFI structure types. While both ZSM-5

and silicalite share the same structure type, they are otherwise distinctly different molecular sieves. ZSM-5 is a zeolite. Silicalite is not a zeolite but instead is a silica polymorph. Attached hereto as Exhibit C is a copy of the Flanigen et al article ("Flanigen, E. M., et al, "Silicalite, a new hydrophobic crystalline silica molecular sieve," *Nature*, Vol. 271, February 9, 1978) referenced in page 93 of Meier et al, which together with U. S. Patent No. 4,061,724, referenced in Appellants' specification at page 2, provides a description of silicalite.

The effect of minimizing the diene content of the feedstock, specifically by a preliminary hydrogenation procedure, is illustrated by Example 15, with results shown in figure 7, and Comparative Example 4, with results shown in figure 8 (pages 39 and 40). In each of Example 15 and Comparative Example 4, dealuminated silicalite, having a silicon/aluminum atomic ratio of 182 in Example 15 and 180 in Comparative Example 4 is employed in the olefin cracking procedure. In Example 15, the feedstock was a light crack naphtha containing 37 wt. % olefins which was pretreated in order to hydrogenate the dienes originally present. In Comparative Example 4, employing a catalyst from Example 4 (page 29), the silicalite catalyst had a silicon/aluminum atomic ratio of 180. The feedstock had not been subject to a preliminary hydrogenation procedure, and had a diene content of 0.5 wt. %. In Example 15 (reported in figure 7), the inlet temperature was 557°C, and the space velocity (LHSV) was 25 h⁻¹. In Comparative Example 4, the inlet temperature was 570°C and the LHSV was 27 h⁻¹. As can be seen from an examination of figure 7, the propylene yield, after the initial startup of the process remained relatively constant over the life of the process up to about 220 hours onstream. In contrast, as shown in figure 8, Comparative Example 4 which had a diene content of 0.5 produced a steeply declining propylene yield from an initial value of about 18 wt. % to about 6 wt. % after about 140 hours onstream.

As illustrated by the experimental work and as described in appellants' specification, at page 40 the feedstock containing a relatively high diene content resulted in a substantial and rapid deactivation of the catalyst. However, by initially hydrogenating the feedstock to hydrogenate the diene content therein, the result was a prolonged period of catalyst stability of up to 220 hours onstream.

Prior Art

The prior art relied upon in rejecting claim 37 (formerly claim 16) under 35 USC 102(e) is U.S. Patent No. 6090271 to Carpeny et al.

The prior art relied reference, upon in rejecting the claims under 35 USC 103 are EP109060 and U.S. Patent No. 5,306,852 to Cosyns et al.

Issues

The issues in this appeal are:

1. whether Claim 37 (originally dependent claim 16) is anticipated by Carpeny; and
2. claims 37 and 17-20 are obvious in view of EP109060 combined with Patent No. 5,306,852 to Cosyns.

Grouping of Claims

In the argument presented in this Brief, all of the claims do not stand and fall together. In addition to the arguments presented with respect to independent claim 37, additional arguments for patentability are presented with respect to claim 20 and to claims 17, 18 and 19, dependent from claim 37.

Appellants' Arguments

Turning first to the rejection of claim 37 (originally claim 16) as anticipated by Carpeny, appellants would respectfully submit that Carpeny fails to anticipate this claim for the following reasons:

1. Carpeny fails to disclose a feedstock having a maximum diene concentration of 0.1 wt. %;
2. Carpeny fails to disclose the use of an MFI type of catalyst having a silicon aluminum atomic ratio of 180-1,000, and a reactor at an inlet temperature of 500-600°C in conjunction with a space velocity of 10-30 h⁻¹; and
3. Carpeny fails to disclose that the feedstock and effluent have substantially the same olefin content by weight with the effluent having a higher propylene content.

With respect to the first point raised above, the feedstock in Carpeny has a diene content far in excess of the 0.1 wt. % maximum called for in appellants' claim 37. In Carpeny, dienes are actually added as necessary to bring the diene content to a level of at least 2%, some 20 times the maximum diene content permitted in appellants' process as set forth in claim 37. Thus as specified in column 4, lines 48-50 of Carpeny, the diolefin content should be within the range of 2-50 wt. % and preferably in the range of 10-20%.

Carpeny does not disclose the use of an MFI type silicate having a silica/aluminum ratio of 180-1,000. Carpeny discloses a very broad silica/aluminum ratio with respect to the wide variety of catalysts disclosed in Carpeny which, with the exception of ZSM-5, are not MFI silicates. Where ZSM-5 is to be employed, Carpeny specifically states that the ZSM-5 as

described in Patent No. 3,702,886 to Argauer is to be employed. Argauer states that the ZSM-5 disclosed there has a much lower silicon/aluminum ratio than the minimum ratio of 180 called for in appellants' claims. In fact, Argauer appears to disclose a maximum silica/alumina ratio of 100 (corresponding to a silicon/aluminum atomic ratio of 50) and in the various examples discloses even lower silicon/aluminum ratios of about 50 or less. Clearly, the only MFI-type silicate disclosed in Carpency has a silicon/aluminum ratio far below the 180-1,000 ratio called for in claim 37.

Further, the patent to Carpency fails to disclose passing the feedstock over the catalyst at a space velocity of $10-30 \text{ h}^{-1}$ at an inlet temperature of $500-600^{\circ}\text{C}$. Carpency discloses a very broad range of space velocities in conjunction with an inlet temperature ranging up to 750°C . The preferred temperature is said to be at $575-625^{\circ}\text{C}$, and the examples show that when operating at or near this temperature range, the space velocity should be relatively low. In fact, the various examples in Carpency show space velocities on the order of 1 h^{-1} WHSV. This, of course, is far below appellants' space velocity (LHSV) of from $10-30 \text{ h}^{-1}$.

Thus Carpency discloses a diene content in the feed far in excess of that specified in claim 37 and further to the extent an MFI silicate (ZSM-5) is involved, discloses a silicon/aluminum atomic ratio well below the minimum called for in appellants' claim 37. Further, if Carpency operates at a temperature within the range called for in appellants' claim 37, the space velocity in Carpency is well below the LHSV of $10-30 \text{ h}^{-1}$ as specified in claim 37. It is clear that the reference fails to anticipate appellants' claim 37.

Turning now to the rejection of claims 37 and 17-20 as unpatentable over EP '060 in view of Cosyns et al, applicants would respectfully submit that the teachings of EP '060 and Cosyns

cannot be selectively combined as proposed in the Final Rejection without a hindsight reconstruction of the prior art references made with the benefit of appellants' disclosure. Further, even if one of ordinary skill in the art were to attempt to combine EP '060 and Cosyns, the results clearly would not involve the selective hydrogenation of dienes in an olefin-rich stream to form at least one olefin and further to form a feedstock having a maximum diene concentration of 0.1 wt.%. In this respect, Cosyns does not disclose the selective hydrogenation of a feedstock even remotely similar to that involved in EP '060 or in applicants' process. The feedstock in Cosyns is not an olefin-rich stream, but instead is a feedstock containing high concentrations of aromatic compounds. The only analysis of the feedstock employed in Cosyns is disclosed in the first table appearing in column 4 of the reference. This table shows that the feedstock is 68 wt. % aromatics, followed by 16% dienes, 12% paraffins, and only 4% olefins. Appellants would respectfully submit that a bare similarity in feedstocks between the two references would not, in itself, provide a basis for combining the prior art references, but here even that factor is clearly missing. The feedstocks of the references are vastly dissimilar. Further, Cosyns does not disclose that the dienes are converted into olefins, and it is clear that Cosyns does not suggest a maximum diene concentration of 0.1 wt.%. In fact, if the Cosyns procedure is somehow combined with EP '060, the various maleic anhydride index (MAI) values given in Cosyns confirm that the diene content for the resulting hydrogenated product would be much higher than the maximum diene content specified in applicants' claims. In this respect, the various examples in Cosyns disclose MAI values ranging from 2-8 for the hydrogenated product. Based upon an initial MAI value of 106 for the feedstock containing 16% dienes, it is evident that the diene content of the product may range from about 3 up to about 10 times applicants' maximum diene content of 0.1%. Specifically based upon the initial

MAI of 106 for a diene content of 16 wt. % dienes, the diene content, based on the outlet MAI would appear to range from about 0.3 (for an MAI outlet value of 2) to 1.2 for an MAI outlet value of 8. It further will be noted that Cosyns even if combined with the teachings of EP109060 would not lead to a diene hydrogenation procedure carried out at a liquid hourly space velocity within the range of 2-5 h⁻¹. In fact, Cosyns does not appear to address the subject of space velocity at all.

Further, with respect to the combination of EP 109 060 and Cosyns, the basic reference does not disclose the range of the silicon/aluminum atomic ratio of 180-1000 as called for in independent claims 20 and 37. In fact, EP '060 not only fails to disclose this range, but instead simply refers to a silicon/aluminum ratio of at least 175 with no upper limit. What is actually disclosed is a catalyst in which no aluminum is present, *i.e.* a silicon/aluminum ratio of infinity. Thus, the teaching in EP '060 is that the silicon/aluminum atomic ratio is of no significance and, in fact, that it makes no difference whether or not aluminum is even present in the catalyst. To the extent that the rejection based on EPO 060 and Carpency relies upon the premise that the disclosure of a very broad range encompassing a narrow range in itself amounts to a disclosure of the narrow range or renders the use of the narrow range obvious, appellants would respectfully refer to the following decisions which hold the existence in the prior art of a broad range which encompasses a narrow range presented in the claims does not in itself establish lack of novelty or obviousness of such ranges. In the decision in *In re Russell*, 169 USPQ 426, CCPA (1971) the court in reversing the rejection of the claims stated:

Essentially, appellant's contention is that the employment of the proportions recited in the claims unexpectedly yields clear compositions without the need for a filtration step. Appellant's position on the law is sound, for even though part of appellant's range of proportions, and all of his ingredients, are suggested by

the broad teaching of Wei, if appellant can establish that his relatively narrow ranges yield unexpectedly superior results as against the broad Wei ranges as a whole, appellant will have established unobviousness of the claimed invention. See *In re Luvisi*, 51 CCPA 1063, 342 F.2d 102, 144 USPQ 646 (1965); *In re Neave*, 54 CCPA 999, 370 F.2d 961, 152 USPQ 274 (1967). p. 428

For a similar holding, attention is respectfully invited to *In re Waymouth and Koury*, 182 USPQ 290, CCPA (1974). In this case, the claims on appeal were directed to a lamp having an arc tube containing halogen in mercury atoms present in a ratio of 0.08 to 0.75. The prior art reference disclosed a similar device containing halogen and mercury atoms. The calculated ratio of halogen to mercury atoms inherently disclosed in the reference ranged from 0.0000001 to 1.3. Although the prior art range enveloped the narrower range claimed by the appellant, the court reversed the rejection of the claims in view of the unexpectedly superior results achieved by operating within the claimed range. In the present case, similarly as in the *Russell* and *Waymouth* decisions, appellants' invention involves the use of the catalyst having characteristics which are not taught by the prior art and which produce unexpectedly superior results which are not recognized by the prior art references.

In addition to the requirement in claims 20 and 37 of a silicon/aluminum atomic ratio of 180 to 1,000, these claims require an inlet temperature of 500° to 600° C and a space velocity (LHSV) of 10 to 30 hrs⁻¹ (claim 37) or an olefin partial pressure of from 0.1 to 2 bar (claim 20). EPA '060 does not disclose or suggest these particular combinations of parameters and instead discloses, in addition to the very broad range of the silicon/aluminum ratio, broad ranges of olefin partial pressure, space velocity, and temperature. Thus, EPA '060 specifies a temperature of 400° to 600° C, a space velocity of 5 to 200 hrs⁻¹, and a pressure of 1.5 to 7.5 atmospheres.

The only qualification on space velocity appears to be a space velocity of less than 50 hrs⁻¹ if the pressure is atmospheric and greater than 50 hrs⁻¹ at a pressure of from 1.5 to 7.5 atmospheres. In fact, with the single exception of Example 36, the space velocity in EPA '060 is well below the 10 to 30 hrs⁻¹ range called for in claim 37 or well above this range as in Examples 32-35. Example 36 of EP '060 does disclose a space velocity of 20 hrs⁻¹. However, Example 36 does not specify a silicon/aluminum atomic ratio within the range called for in appellants' claims, and the only inference that can be drawn from the reference disclosure is that this ratio, like the ratio of the other examples of silicalite -1 disclosed in the reference, is well above the upper limit of 1,000 called for in the claim. In this respect, every example of silicalite -1, as described in EP '060 specifies that no aluminum is present.

Further with respect to claim 37, appellants would respectfully submit that the Examiner is in error in the apparent position that the various examples in EP '060 disclose that the olefin contents of the feed and product are substantially the same, or within $\pm 15\%$ of each other. As to the relative olefin content of the feed and the effluent, there is no clear indication in the various examples that these values are within the $\pm 15\%$ of each other as alleged in the Final Rejection. In some cases they are clearly outside of this range. For example, the products in Examples 16-23, which show high selectivity to BTX, clearly do not involve an effluent having an olefin content within 15% of the olefin content of the product stream. In other examples it is impossible to determine whether the claimed relationship between the feed stream and the effluent stream is met. Similarly, Examples 1-6 show a very high product in the effluent of C₅₊ compounds. It is not clear whether these are saturated or unsaturated, and thus again, a determination as to the relevant olefin content of the feedstock and effluent cannot be made. In

any event, it will be recalled that whether or not these various examples involve an effluent and a feedstock having olefin contents within ± 15 , they clearly do not involve the space velocity requirements of independent claim 37. The sole example of EP '060, which does indicate a space velocity within applicants' claimed range is Example 36. Example 36 does not, however, disclose a relationship between the olefin content of the feed and the effluent of $\pm 15\%$. In fact, from what is disclosed in Example 36, the relative olefin content of the feed and effluent could well be far outside this range. In this respect, the olefin content of the feed in Example 36 is 100% normal butene whereas the identified olefin content of the effluent (propylene, ethylene, and isobutylene) is about 50%. About 4-5% of the effluent is identified as saturated gases, with the remainder being identified as C₅₊ liquids. Presumably, substantial portions of these liquids would be olefins resulting from butene oligomers which are unsaturated. However, if as much as even 30% of the C₅₊ liquids are saturated, it is clear that the olefin content of the effluent would not be within 15% of the olefin content of the feedstock as alleged in the Final Rejection.

As to the Examiner's alternative ground based upon inherency, appellants would respectfully note that the law is well settled that for an alleged inherent feature to result from a prior art teaching, it is necessary that this inherent feature necessarily flow from the teachings of the prior art. Thus, an alleged inherent feature must be a necessary result and not merely a possible result. This principle is stated in MPEP Section 2112:

The fact that a certain result or characteristic may occur or be present in the prior art is not sufficient to establish the inherency of that result or characteristic. (Emphasis original)

Section 2112 reflects the general rule that for inherency to reside, it must be shown that the alleged inherency is necessarily present and not a mere possibility. Thus, as stated by the Board in *Ex parte Keith*, 154 USPQ 321 (Bd. of App. 1966), in reversing the Examiner's rejection based upon inherency:

There are other possible courses the reaction could follow . . .
Asserted inherency must be a necessary result and not merely a possible result.

As indicated in Section 2112, this principle was more recently followed by the Board in *Ex parte Levy*, 17 USPQ2d 1461 (Bd. of App. and Interf. 1990), where the Board reversed an inherency rejection, stating as follows:

In relying upon the theory of inherency, the examiner must provide a basis in fact and/or technical reasoning to reasonably support the determination that the alleged inherent characteristic necessarily flows from the teachings of the prior art (citing cases). (emphasis original).

Here, this claimed subject matter clearly is not inherent in EP '060.

As demonstrated above, the combination of Cosyns with EPO 060 cannot be made in a manner to arrive at appellants' invention even if it were appropriate to use appellants' teachings as a basis for combining the diverse teachings of the references. However, as the Board will recognize, this is not the appropriate standard to be applied in combining prior art teachings. This standard forbids using appellants' own disclosure as a basis for assembling prior art teachings and requires a suggestion or motivation in the prior art as reflected in decisions in *ACS Hospital Systems, Inc. v. Montefiore Hospital*, 221 USPQ 929 (Fed. Cir. 1984) and *Ex parte*

Giles 228 USPQ 886 (PTO Bd. Of Appeal. And Int. 1985). As expressed by the Board in *Giles* at 688:

Only appellant's disclosure and not the prior art provides a motive for achieving the combination as claimed by the appellant. To imbue one of ordinary skill in the art with knowledge of the invention . . . when no prior art reference or references of record convey or suggests that knowledge is to fall victim to the insidious effect of a hindsight syndrome wherein that which only the inventor taught is used against its teacher.

Attention is also respectfully invited to the Federal Circuit decision in *In re Fine*, 5 USPQ2d 1956 (Fed. Cir. 1988) wherein the Court stated at 1600:

It is essential that "the decisionmaker forget what he or she has been taught at trial about the claimed invention and cast the mind back to the time the invention was made . . . to occupy the mind of one skilled in the art who is presented only with the references, and who is normally guided by the then-accepted wisdom in the art." *Id.* One cannot use hindsight reconstruction to pick and choose among isolated disclosures in the prior art to deprecate the claimed invention. (Emphasis added)

The admonitions in *Ex parte Giles* and *In re Fine* are to step back in time to evaluate the invention only in the context of what is disclosed in the references without regard to what is disclosed in appellants' specification. When this is done, it is believed clear that one of ordinary skill in the art would not arrive at appellants' invention based only upon the references and "the then-accepted wisdom in the art."

The Final Rejection offers no motivation in the prior art references for combining the teachings of EP 060 and Cosyns. The argument that the EP 060 does not require the presence of dienes ignores the fact that EP 060 does not even address the subject of dienes in the feedstock.

There is no disclosure in EP 060 that dienes should not be present in the feedstock, and no motivation for one skilled to look to a process suitable for diene removal. The motivation for diene removal is not found in EP 060, it is found, as a note above, only in appellants' disclosure.

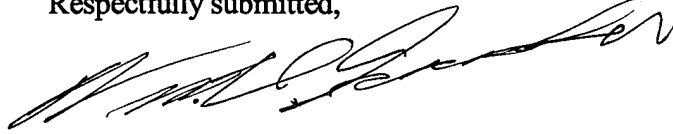
For the reasons advanced above, it is respectfully submitted that this application is in condition for allowance, subject only to the possibility of a double-patenting rejection should an issue presented by the provisional double-patenting rejections come into being. As indicated previously in the prosecution of this application, a terminal disclaimer will be filed when timely and appropriate.

Conclusion

For the reasons set forth above, Appellants respectfully submit that all of the claims herein are patentable over the prior art. Accordingly, it is respectfully requested that the Final Rejection of the claims be reversed.

A check in the amount of \$320.00 is enclosed for the statutory fee. The Commissioner is hereby authorized to charge our Deposit Account No. 12-1781 for any deficiency in fees connected with this communication or to credit any overpayment.

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'W.D. Jackson', written in a cursive style.

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Date: December 11, 2002

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